

European Patent No. 0 848 294
European Patent Application No. 97203898.8
DSM N.V., Japan synthetic Rubber Co. Ltd., and Japan Fine Coatings Co. Ltd.
Opposition by Vantico AG

# Statement of Grounds of Opposition including Facts and Arguments in Support

## 1. Grounds of Opposition

The subject matter of European Patent No. 0 848 294 (the Patent) is not patentable (i) because it is not new and does not involve an inventive step; and (ii) the Patent does not disclose the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art.

## 2. Requests

The Opponents request that the Patent be revoked in its entirety. In the event that the Patent is not revoked in its entirety, the Opponents request Oral Proceedings.

### 3. The Patent

The Patent relates to a process for photo-fabricating a three-dimensional object comprising selectively curing a defined photo-curable resin composition containing an oxetane compound, an epoxy compound and a cationic photo-initiator. Claims 1 and 2 are independent claims; in claim 1, the oxetane compound is a compound comprising two or more oxetane rings; in claim 2, the oxetane compound has one or more groups according to the formula

wherein Z is oxygen or sulphur and wherein  $R^1$  and  $R^2$  constitute the remainder of a molecule.



Claims 3 to 9 further specify the composition to be used in the process of the invention. Claims 10 to 13 specify certain physical properties which must be possessed by the composition when cured.

Claim 14 is an independent claim which reads "A three-dimensional object comprising a cured photo-curable resin composition according to any one of claims 1-13."

# Priority Date of the Patent

None of the claims of the patent are entitled to the claimed priority date.

We refer to the English language translation of JP 35289396 (the priority application for the Patent) on the file of the Patent. Page 55 and 56 (claim 1 and the abstract) disclose a photo-curable resin comprising (A) a compound having an oxetane ring, (B) a compound having an epoxy group, and (C) a cationic photo-initiator. However, the description at page 19 lines 6 to 12, states "The compound having an epoxy group (hereinafter may be called from time to time component (B)) which is the component of the resin composition of the present invention is a compound having an epoxy group and a number average molecular weight reduced to polystyrene of 1,000-20,000, preferably 1,500-10,000, and more preferably 2,000-5,000, measured using gel permeation chromatography." (our emphasis). That the draftsman intended to limit the scope of the invention to compositions in which the epoxy compound had the specified molecular weight, is further confirmed by the presence of comparative Example 3 in Table 1 of the priority document. The composition of this Example does not contain a compound having an epoxy group having a number average molecular weight reduced to polystyrene of 1,000-20,000 measured by gel permeation chromatography, but it does contain a different class of epoxy compound. The priority document clearly indicates that the composition of comparative Example 3 is not regarded as being within the scope of the invention: "...the tensile elongation of the resin compositions excluding the component (B) which were prepared in the comparative Examples 1 and 3 was 5% and 7% respectively. Therefore, the toughness of the resin compositions excluding the component (B) was insufficient for photo-fabricating applications." (lines 1 to 6 page 53). See also page 53 line 2 from the bottom to page 54 line 3, "...the fabricated products produced from the comparative Examples 3 and 5 exhibited large dimensional differences so that sufficient dimensional accuracy required for photo-fabrication could not be obtained."

The only disclosure in the priority document of compositions containing the generalised class of epoxy compounds is that in claim 1 and the abstract. However, that disclosure is in combination with the broad class for component (A), "a compound having an oxetane ring". Claims 1 and 2 of the Patent do not have component (A) defined in this way; for claim 1, component (A) is an oxetane having two or more oxetane rings, while for claim 2, the oxetane has a defined formula in which R<sup>1</sup> and R<sup>2</sup> "constitute the remainder of a molecule". There is no disclosure in the priority document of the combination of either of these defined components (A), with component (B) defined as an epoxy compound.



Accordingly claims 1 and 2 are not entitled to the priority date. Similar reasoning applies to all the remaining claims except for claim 5.

Claim 5 specifies that the epoxy compound has a number average molecular weight of 1,000-20,000 but does not specify that this number is reduced to polystyrene and measured by gel permeation chromatography. Claim 5 is therefore not entitled to its priority for this reason. Even if claim 5 were to be amended to agree with the disclosure of the priority document, it would not be entitled to its priority date insofar as it is dependent on claim 2, for the reasons given in the following paragraph.

In addition, the words "in which  $R^1$  and  $R^2$  constitute the remainder of a molecule", or anything similar, do not appear anywhere in the priority document. Rather,  $R^1$  and  $R^2$  are defined in the lengthy passage bridging pages 6 and 7. Accordingly, claim 2 is not entitled to the priority date for this additional reason. Similar reasoning applies to claims 5 to 14 insofar as they are dependent on claim 2.

In summary, the claims of the Patent are not entitled to the priority date of 13<sup>th</sup> December 1996, and should be accorded the date of filing of the application, 12<sup>th</sup> December 1997.

### 4. The Prior Art

### (a) Prior Art under Article 54(3) EPC

# D1. EP 831373A, priority date 20th September 1996 (DSM et al.)

This European application has a priority date of 20<sup>th</sup> September 1996, and a filing date of 19<sup>th</sup> September 1997. It therefore forms part of the state of the art under Article 54(3) EPC.

The document discloses compositions which comprise (A) a compound having a cyclohexene oxide structure; (B) a cationic photo-initiator; (C) an ethylenically unsaturated polymer; (D) a radical photo-initiator; and (E) spherical silica particles (claim 1 and line 57 page 2 to line 5 page 3). Component (A) is an epoxy compound. In addition to the essential components (A) to (E), other components may be incorporated (line 6 page 8). Specific examples of such optional components include oxetanes such as trimethylene oxide, 3,3-dimethyl oxetane, 3,3-dicholoromethyl oxetane, 3-ethyl-3-phenoxymethyl oxetane, and bis(3-ethyl-3-methyloxy)butane (lines 37 to 39 page 8). 3-Ethyl-3-phenoxymethyl oxetane is an oxetane of the formula specified in claim 2 of the patent in suit. The compositions are used for the manufacture of three-dimensional objects by selective irradiation using visible light, ultraviolet light or infrared light (lines 41 to 47 page 9).

EP <u>837366A</u> (priority 18<sup>th</sup> October 1996); EP <u>848292</u>A (priority 10<sup>th</sup> December 1996); and EP <u>848293</u>A (priority 10<sup>th</sup> December 1996) all have disclosures very similar to that of EP 831373A.



## (b) Prior Art under Article 54(2) EPC

## (i) Prior art relating to Photo-curable Compositions

Photo-curable compositions as defined in the claims of the Patent are known. In addition to prior art cited during prosecution of the patent in suit, the following documents are examples of prior art disclosures of such compositions.

# D2. JP 8-269392, published 15th October 1996 (Toagosei Chemical Industry Co.)

An English translation of this document is enclosed. The document states "...the first invention of the present application is an energy-beam-curable composition used as a primary coating for optical fibers composed of a compound having 1-4 oxetane rings and a photocationic polymerization initiator. The second invention is the energy-beam-curable composition used as a primary coating for optical fibers specified in the first invention, wherein a compound containing an epoxy group also is included." (paragraph 0005, lines 7-12). Preferred oxetanes are listed on pages 8 onwards, and include many of the compounds used in the Patent, see for example formulae 1, 2, 9 and 10. Preferred epoxy compounds are listed on pages 25 onwards. Specific examples of compositions containing an oxetane and an epoxy are Examples 5 and 7 in Table I of the Examples. Here, the oxetane is of formula 32:

and the epoxy is of formula 36:

$$0 \longrightarrow -\overset{0}{\text{C}} - 0 - \overset{0}{\text{C}} - 0 - \overset{0}{\text{C}} \longrightarrow 0$$

$$(36)$$



### D3. US 3835003, published 1974 (American Can Co.)

This document shows that compositions containing an oxetane, an epoxy and an initiator were known at least as long ago as 1974. Specific oxetanes mentioned which fall within the definitions of the Patent are 3,3-bis(methoxymethyl)oxetane, 3,3-bis(phenoxymethyl)oxetane and 3,3-bis(acetoxymethyl)oxetane (lines 7-9 column 3). Example 4 describes a mixture of 3,3-bis(chloromethyl)oxetane with glycidyl methacrylate-allylglycidyl ether copolymer. Column 8 describes the use of the compositions as follows: "The polymers produced by the polymerization process of the present invention when [oxetane] monomer and epoxides are employed are useful in a wide variety of applications in the field of graphic arts, due to their superior adhesion to metal surfaces, excellent resistance to most solvents and chemicals, and capability of forming high resolution images. Among such uses are photoresists for chemical milling, gravure images, offset plates, stencil-making, micro-images for printed circuitry, thermoset vesicular images, micro-images for information storage, decoration of paper, glass, and packages, encapsulation of printed circuits, and light-cured coatings."

# D4. US 5674922, published 7th October 1997 (Igarashi et al)

This document discloses active energy beam-curable compositions which comprise (1) at least one compound having one oxetane ring and one hydroxyl group in the molecule, (2) at least one compound having one or more oxirane rings in the molecule and (3) at least one compound which initiates cathionic polymerization by irra. ation of active energy beams. (abstract, claim 1). Preferred oxetane compounds are those of formula (I):

$$R_1$$
  $R_2$  OH (I)

wherein  $R_1$  represents a hydrogen atom, alkyl group having 1 to 6 carbon atoms, fluoroalkyl group having 1 to 6 carbon atoms, allyl group, aryl group, furyl group or thienyl group; and R2 represents an alkylene group having 1 to 6 carbon atoms or an alkylene group containing an ether linkage (lines 43-60 column 2).  $R_2$  is preferably a methylene group (lines 62-63 column 2. Example 1 discloses a composition comprising 3-ethyl-3-hydroxymethyloxetane together with the diglycidyl ether of Bisphenol A (an epoxy compound) and a photo-initiator. Examples 2 and 3 disclose mixtures of the same oxetane with the epoxy compound of formula

$$0 \longrightarrow -C - C - C \longrightarrow 0$$





The compositions have a variety of uses, including paints for wood-workings, paints for plastics, paints for metals, paints for papers, luster varnish, coatings for protection, decoration and insulation, paints for optical fibers, injection sealing compounds, printing inks, sealants, adhesives, photoresists, wire-insulating materials, textile coatings, laminates, impregnated tapes, printed plates, etc. (paragraph bridging columns 8 and 9).

# D5. J. Polymer Sci. Part A, Polymer Chemistry, 33, 1807-1816 (1995)

This document is entitled "Photoinitiated Cationic Polymerization of Oxetane Formulated with Oxirane". It is a detailed study of the polymerisation mechanisms involved in the polymerisation of oxetanes and oxiranes (epoxies) separately and together. Two oxetanes, both of the type used in the Patent, and two epoxies, were studied, using diphenyl-4-thiophenoxyphenyl sulphonium hexafluoroantimonate as a cationic photoinitiator (bottom of column 1 page 1808).

# (ii) Prior Art relating to the Photo-fabrication of Three-dimensional Objects

The photo-fabrication of three-dimensional objects comprising selectively curing a photo-curable composition is well known. For example:

### D6. EP 535 828 A, published 1993 (ICI)

This document describes techniques previously known in the field of photo-fabrication, and sets out to solve the problem that the cure depth of the compositions used have previously been greater than 500 microns. It does this by applying light of a defined wavelength to a composition containing a mix of free radical and cationic polymerisable compounds and a dyestuff with particular absorption characteristics. The compositions may contain epoxies; many epoxies are listed at lines 21-55 page 3, while at line 35 page 5 to line 5 page 6 it is stated "Of the compounds capable of cationic polymerisation, known aromatic, alicyclic or aliphatic epoxy compounds having at least two epoxy groups per molecule are preferred", and such epoxies are listed. Oxetanes are mentioned at lines 6 to 8 page 6 as possible constituents of the compositions, trimethylene oxide, 3,3-dimethyloxacyclobutane, and 3,3-bis(chloromethyl)-oxacyclobutane being mentioned as examples, and lines 15 to 16 page 8 state that "The cationic polymerisable compound may be used singly or in mixtures of two or more such compounds to provide the desired properties."

### (iii) General Background Prior Art

# D7. Macromol. Symp. 107, 125-138 (1996)

This document was cited in the European prosecution of the Patent and provides useful background. Its title is "Oxetane Photopolymerization - A System with Low Volume Shrinkage". Lines 6-7 page 129 state "...we consider oxetanes an interesting family of compounds for applications in which reduced shrinkage in volume is important".



### 5. Patentability of the Patent over the Prior Art

### (a) Novelty

For the reasons given above, claims 1 to 4 and 6 to 14 are entitled only to the date of filing of the application, 12<sup>th</sup> December 1997. Document D1 is part of the state of the art under Article 54(3) EPC and discloses all the features of claim 2. Claim 2 therefore lacks novelty.

The same reasoning applies to the three additional documents referred to under section 4(a) above.

For consideration of claim 14, see section 6(c) below.

### (b) Inventive Step

D6, EP 535 828 A, is document D2 relied upon by the European examiner during the prosecution of the Patent. The examiner argued that the document deprived the original claims of novelty, and said "The Applicant is invited to argue, whether in present claim 1 there can be seen a "selection invention" over D2." The Applicant reduced the scope of the oxetane which could be present in the claims to exclude those specifically disclosed in EP 535828A (trimethylene oxide, 3,3-dimethyloxacyclobutane, and 3,3bis(chloromethyl)-oxacyclobutane). In the reply dated 23<sup>rd</sup> November 1999 to the official action, the Applicant argued: "The closest prior art is document D2. D2 relates to stereolithography and provides a three-dimensionally mouldable photocurable resin composition having a cure depth of 0.5mm or less when exposed to visible light (see D2 page 2, lines 27-28). The objective problem between D2 and the present invention, is to provide a three dimensional article very rapidly, while the article has at the same time a high toughness and dimensional accuracy. The present invention solves this problem by providing a process in which certain types of oxetanes are used. None of the cited documents suggest or teach this solution to the skilled man. The solution is not obvious to this skilled man. Therefore claims 1 and 2 involve an inventive step."

The examiner allowed the claims without further action.

If the Applicant is right in his analysis of the prior art, then the burden is on the Applicant to show that his solution to the problem posed by the prior art, i.e. "providing a process in which certain types of oxetanes are used", is indeed inventive. The Applicant has completely failed to do this. No data is present in the Patent, and no data was submitted during prosecution, to show that the use of the particular oxetanes claimed, gives any special or surprising effect.

The examiner correctly appreciated that, in order to be patentable over D6, the invention of the Patent would need to be regarded as a selection invention. In the absence of any



evidence to show that the specific compositions described in the Patent exhibit a surprising effect, the invention must be deemed to be obvious over D6.

The only data provided in the Patent relate to compositions containing a single oxetane, 1,4-bis(3-ethyl-3-oxetanylmethoxy)methylbenzene, Component A in Table 1. No data are given to show that even this one compound, let alone any of the other oxetanes which are specified in the claims of the Patent, gives surprising results over what would be obtained using compositions described in D6.

Moreover, compositions used in the process of the Patent are known in the art, see documents D2, D3, D4 and D5 above. Compound A of Table 1 of the Patent, referred to above, is compound 32 of D2, and mixtures of it with epoxies are known. These mixtures are known to be useful in a number of applications, for example coatings. The Patent states (line 45 page 2) that minimal deformation due to shrinkage during curing with light, is a characteristic required by a composition used for photo-fabrication processes. This is also a key requirement for compositions to be used in coatings, for the obvious reason that shrinkage of a coating would lead to poor adhesion to the substrate. Further, it is specifically known that compositions containing oxetanes are useful in applications where low volume shrinkage is important - see document D7 above. It would have been obvious to try such compositions in the process claimed in the Patent. The results obtained would not have been a surprise to the man skilled in the art.

None of the claims of the Patent provide any inventive solution to the problem of producing improved compositions for the use of photo-fabrication of three-dimensional objects. Claims 1 to 4 define particular oxetanes for use in the process of the Patent. No data is provided showing a surprising effect by using any oxetane included within claims 1 to 4 in comparison with any oxetane outside those claims.

Claim 5 is dependent on claims 1 to 4. It specifies that the epoxy compound should have a particular molecular weight. It is difficult to image that there could be any inventive step in using an epoxy compound of a particular molecular weight, where the chemical structure is otherwise completely unspecified.

Claim 6 relates merely to commonplace formulation design which would be well within the skills of the man skilled in the art. Claims 7 and 8 require the composition also to include a polyol. No evidence is provided that such compositions are inventive.

Claims 10 to 13 are dependent on earlier claims specifying the use of particular compositions, and relate merely to desirable results which the man skilled in the art would wish to obtain when working with photo-curable compositions. It should be noted that Comparative Examples 3, 4 and 5 of the Specification report in Table 2 a Young's modulus of greater than 80, meeting the requirement of claim 10. Comparative Example 5 meets the requirement of claim 11, and Comparative Examples 2 and 5 meet the requirement of claim 12. Comparative Examples 2 and 5 meet the requirement of claim 13.



No inventive step can be seen for claims 10 to 13 in the absence of evidence that the obtaining of such results using the compositions specified in claims 1 to 9 is surprising.

Insofar as Claim 14 has any meaning, it can have no inventive step independent of claims 1 to 13.

In summary, all claims of the Patent lack inventive step.

### 6. The scope of all the claims of the Patent is obscure

## (a) The process claims 1 to 13 are unclear in scope

Claims 1 to 13 claim "a process for photo-fabricating a three-dimensional object comprising selectively curing a photo-curable composition...". This wording is unclear in two respects: the limitation imposed by the words "a three-dimensional object" is unclear, as is the word "selectively". The prior art referred to above discloses photo-curable compositions falling within the definitions of the Patent. When cured, these compositions form or become part of an object. Preparation of such objects as offset plates or stencils (D3) must by definition be by "selective" curing, whatever the intended meaning of "selective".

If the intention of the Patentee is to claim "a photo-fabrication process for forming threedimensional objects consisting of integrally laminated cured resin layers prepared by repeating a step of forming a cured resin layer by selectively irradiating a liquid photocurable material with light" (lines 10-12 page 1 of the Patent) then the claims should be worded accordingly. We have assumed such a wording in our comments on the patentability of the process claims elsewhere in this Statement of Grounds.

### (b) Claims 1 to 4 and 6 to 14 are unclear in scope regarding Component (B)

All claims of the Patent require the presence of an epoxy compound. Apart from claim 5, which specifies that the epoxy compound should have a number average molecular weight of 1,000-20,000, all claims permit any epoxy compound to be used. However, the Examples in the specification are inconsistent with the claims. Table 1 on page 17 lists the components present in five Examples which illustrate the invention, and five comparative Examples. Comparative Example 3 contains an oxetane (1,4-bis(3-ethyl-3-oxetanylmethoxy)methylbenzene) as specified for component (A) in claim 1; an epoxy compound (3,4-epoxycyclohexylmethyl-3',4'-epoxycyclohexane carboxylate), as specified for component (B) of claim 1; and a cationic photo-initiator (UVI-6974) as specified for component (C) of claim 1. Accordingly the use of a composition as specified in comparative Example 3 of the specification appears to fall within the scope of claim 1.

Lines 14 to 17 page 21 of the Specification state: "...the tensile elongation of the resin compositions excluding component (B) which were prepared in the Comparative



Examples 1 and 3 was 5% and 7% respectively. Therefore, the toughness of the resin compositions excluding component (B) was insufficient for photo-fabrication applications"

Lines 29-30 page 21 of the Specification state: "However, the fabricated products produced from the Comparative Examples 3 and 5 exhibited large dimensional differences so that sufficient dimensional accuracy required for photo-fabrication could not be obtained."

Accordingly, comparative Example 3 does not have the utility claimed for the present invention; the composition described cannot be used successfully in the process of the present invention.

We note that in Table 1, the epoxy compound "Epolead PB3600", which is an epoxidated polybutadiene, is labeled as Component B. This is consistent with the passage at lines 14 to 17 page 21 quoted above, and suggests that the draftsman of the Patent intended the scope of the claims to be limited to epoxy compounds of the "Epolead" type as Component B. The claims (other than claim 5) are, however, not so limited.

That the draftsman intended to include only a limited class of epoxy compounds within his claims is further supported by the long section headed "Optional Components" on pages 11 to 14 of the Specification. This section states "In addition to the above essential components (A) to (C), other components may be incorporated into the photo-curable resin composition of the present invention as required to the extent that the effects of this invention are not adversely affected." (lines 42-44 page 11). The section lists a very wide range of optional components, including polyols, monomers such as acrylamide, photosensitizers, diluents and fillers. Crucially, however, it includes the following two passages. "Examples of epoxy compounds other than component (B) are in particular, epoxy compounds with a molecular weight of less than about 1000 which include alicyclic epoxy compounds such as...." (line 51 page 11 to line 13 page 12); and "Other additives include polymers or oligomers, such as epoxy resins other than the abovementioned compound having an epoxy group used as component (B) and other epoxy compounds..." (lines 13 to 14 page 14).

Finally, we note that claim 6 of the Specification as filed was deleted without comment by the Applicants during prosecution of the Patent. This claim specified a composition containing an epoxy compound with a number average molecular weight of less than 1,000.

Our submission is as follows. If Component (B) specified in the claims is intended to include epoxy compounds such as the one used in comparative Example 3, then the invention includes, by the patentees' own admission, unworkable embodiments, and therefore lacks inventive step. If the scope of the claims is intended to be limited only to certain epoxy compounds, then the scope of all claims other than claim 5 is obscure. In either event the Patent does not disclose the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art.



### (c) Claim 5 is indefinite in scope

Claim 5 specifies that the epoxy compound has a number average molecular weight of 1,000-20,000. However, the description (lines 35-38) states that the epoxy compound has "a number average molecular weight reduced to polystyrene of 1,000-20,000 measured using gel permeation chromatography". For high molecular weight materials, the basis on which the molecular weight is measured is important, and must be specified. It should therefore be included in the claims.

### (d) Claim 14 Cannot be Construed

Claim 14 reads "A three-dimensional object comprising a cured photo-curable resin composition according to any one of claims 1-13."

This claim cannot be construed, because claims 1 to 13 do not claim compositions. Rather, they claim a process for photo-fabricating a three-dimensional object which comprises "selectively" curing a composition. If it is the intention of the patentees to claim a three-dimensional object which has been made by such a process, then the claim is unnecessary (Article 64(2)EPC). If, as it appears, the claim is intended to be a claim to a three-dimensional object made by any process from a cured resin composition as defined in any one of claims 1 to 13, then the claim plainly lacks novelty and/or inventive step because the compositions themselves lack novelty and/or inventive step over documents D1, D2, D3, D4 and D5. In either event claim 14 cannot stand and should be deleted.

Our comments on the lack of clarity of the process claims (section 6(a) above) also apply to claim 14. "A three dimensional object" is not a real limitation as, in the real world, all objects are three-dimensional, including those which are described in the prior art documents D2, D3, D4 and D5 above.

#### 7. Conclusions

All the claims of the Patent lack novelty and/or inventive step over the prior art. In addition, all the claims are defective because the Patent does not disclose the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art. The Patent should therefore be revoked in its entirety.

5.1.50.

SCOTT, Susan Margaret Abel & Imray, London. 28<sup>th</sup> May 2002.